

# Order Parameter to Characterize Valence-Bond-Solid States in Quantum Spin Chains

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We propose an order parameter to characterize valence-bond-solid (VBS) states in quantum spin chains, given by the ground-state expectation value of a unitary operator appearing in the Lieb-Schultz-Mattis argument. We show that the order parameter changes the sign according to the number of valence bonds (broken valence bonds) at the boundary for periodic (open) systems. This allows us to determine the phase transition point in between different VBS states. We demonstrate this theory in the successive dimerization transitions of the bond-alternating Heisenberg chains, using the quantum Monte Carlo method.

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About two decades ago, Haldane made a striking conjecture that the integer-spin  $S$  antiferromagnetic Heisenberg chain has a unique disordered ground state with an energy gap, while for a half-odd-integer spin there is no energy gap and the system belongs to the same universality class as the  $S = 1/2$  case [1]. For  $S = 1$ , this conjecture was confirmed by many numerical [2] and experimental studies [3]. Affleck, Kennedy, Lieb, and Tasaki studied an  $S = 1$  isotropic spin chain with special bi-quadratic interactions which has the exact ground state with a finite gap [4]. They also proposed the valence-bond-solid (VBS) state for the Haldane gap systems, and concluded that the ground state of the Heisenberg chain is described approximately by the VBS state. Since the spin configurations of the VBS state show the hidden antiferromagnetic order, den Nijs and Rommelse proposed the string order parameter to characterize the  $S = 1$  Haldane phase [5],

$$\mathcal{O}_{\text{string}}^{\alpha} = - \lim_{|k-l| \rightarrow \infty} \langle \Psi_0 | S_k^{\alpha} \exp \left[ i\pi \sum_{j=k+1}^{l-1} S_j^{\alpha} \right] S_l^{\alpha} | \Psi_0 \rangle, \quad (1)$$

where  $\alpha = x, y, z$  and  $|\Psi_0\rangle$  means the ground state. Thus this order parameter enables us to detect the VBS state *indirectly*. The string order parameter was generalized to  $S > 1$  cases by Oshikawa [6].

On the other hand, Haldane's prediction was also discussed by the Lieb-Schultz-Mattis (LSM) type argument [7]. Using this technique, Affleck and Lieb examined Haldane's conjecture [8]. Later, Oshikawa, Yamanaka, and Affleck generalized this argument, and obtained a necessary condition for a gapped state [9]. However, the relation between the LSM argument and the VBS picture including the string order parameter has not been fully understood. In this Letter, we discuss this relation, and show that an overlap integral appearing in the LSM argument [Eq. (3) below] plays a role of an order parameter which detects VBS ground states *directly*. We

also show that it can also be applied to determine phase boundaries in between different VBS states. We demonstrate this idea by the quantum Monte Carlo (QMC) simulation for successive dimerization transitions in the bond-alternating Heisenberg spin chains.

First, let us review the LSM argument briefly based on Ref. [9] and introduce the order parameter. We consider a periodic spin chain of length  $L$  with short range interactions. We assume the translational ( $\mathcal{T} : S_j^{\alpha} \rightarrow S_{j+1}^{\alpha}$ ) and the parity ( $\mathcal{P} : S_j^{\alpha} \rightarrow S_{L+1-j}^{\alpha}$ ) invariance. Now we introduce the following "twist operator"

$$U \equiv \exp \left[ i \frac{2\pi}{L} \sum_{j=1}^L j S_j^z \right]. \quad (2)$$

Since this operator rotates all the spins about the  $z$ -axis with relative rotation angle between the neighboring spins  $2\pi/L$ , it creates spin-wave-like excitations. Applying the twist operator  $q$  times to the unique normalized ground state  $|\Psi_0\rangle$  generates a set of low-lying excited states  $\{|\Psi_k\rangle \equiv U^k |\Psi_0\rangle\}$  ( $k = 1, \dots, q$ ). The excitation energy of the state  $|\Psi_1\rangle$  is evaluated as  $\Delta E \sim \mathcal{O}(L^{-1})$ . Although  $|\Psi_1\rangle$  is not necessarily an eigenstate of the Hamiltonian, there exists at least one eigenstate with energy of  $\mathcal{O}(L^{-1})$ , if the state  $|\Psi_1\rangle$  is orthogonal to the ground state  $|\Psi_0\rangle$ .

In order to consider the orthogonality of these states, we introduce the following overlap integral which plays a central role in this Letter:

$$z_L^{(q)} \equiv \langle \Psi_0 | \Psi_q \rangle = \langle \Psi_0 | U^q | \Psi_0 \rangle. \quad (3)$$

The invariance under transformations  $\mathcal{T}$  and  $\mathcal{P}$  yields

$$z_L^{(q)} = \langle \Psi_0 | \mathcal{T} U^q \mathcal{T}^{-1} | \Psi_0 \rangle = e^{i2q(S_1^z - m)\pi} z_L^{(q)}, \quad (4)$$

$$= \langle \Psi_0 | \mathcal{P} U^q \mathcal{P} | \Psi_0 \rangle = e^{i2qm\pi} [z_L^{(q)}]^*, \quad (5)$$

where  $m$  is the magnetization per site. Eq. (4) shows that  $z_{\infty}^{(1)} \neq 0$  is possible only when  $S - m = \text{integer}$ .

In this case, the system has a gap without breaking the translational symmetry. On the other hand, for a rational value  $S - m = p/q$  with  $p$  being an integer,  $|\Psi_0\rangle, |\Psi_1\rangle, \dots, |\Psi_{q-1}\rangle$  are mutually orthogonal. This means that the system in the  $L \rightarrow \infty$  limit is gapless ( $z_\infty^{(q)} = 0$ ), otherwise gapped with  $q$ -fold degenerate ground state ( $z_\infty^{(q)} \neq 0$ ) due to the spontaneous breaking of the translational symmetry. Thus  $q(S - m) = \text{integer}$  is a necessary condition for a gapped state [9]. In this case,  $z_L^{(q)}$  is real or pure imaginary due to Eq. (5).

In the previous works based on the LSM argument [7, 8, 9], however, explicit values of  $z_L$  have not been calculated. Now let us evaluate  $z_L$  for various ground states. In the Néel state with two-fold degeneracy, one can obtain  $z_L^{(2)} = (-1)^{2S}$  immediately. In order to calculate  $z_L$  in VBS states, we introduce the Schwinger boson representation for the spin operators [10]:

$$\begin{aligned} S_j^+ &= a_j^\dagger b_j, & S_j^z &= \frac{1}{2}(a_j^\dagger a_j - b_j^\dagger b_j), \\ S_j^- &= b_j^\dagger a_j, & S_j &= \frac{1}{2}(a_j^\dagger a_j + b_j^\dagger b_j), \end{aligned} \quad (6)$$

where these bosons satisfy the commutation relation  $[a_i, a_j^\dagger] = [b_i, b_j^\dagger] = \delta_{ij}$  with all the other commutations vanishing. The operator  $a_j^\dagger$  ( $b_j^\dagger$ ) increases the number of up (down)  $S = 1/2$  variables under symmetrization. Then, a generalized VBS state discussed in Ref. [11] in a periodic system (see Fig. 1(a)) is written as

$$|\Psi_{\text{VBS}}^{(m,n)}\rangle \equiv \frac{1}{\sqrt{\mathcal{N}}} \prod_{k=1}^{L/2} (B_{2k-1,2k}^\dagger)^m (B_{2k,2k+1}^\dagger)^n |\text{vac}\rangle, \quad (7)$$

where  $B_{i,j}^\dagger \equiv a_i^\dagger b_j^\dagger - b_i^\dagger a_j^\dagger$ ,  $\mathcal{N}$  is a normalization factor, and  $|\text{vac}\rangle$  is the vacuum with respect to bosons. The integers  $m$  and  $n$  satisfy  $m + n = 2S$ . Using relations  $U a_j^\dagger U^{-1} = a_j^\dagger e^{i\pi j/L}$  and  $U b_j^\dagger U^{-1} = b_j^\dagger e^{-i\pi j/L}$ , a twisted valence bond  $U B_{j,j+1}^\dagger U^{-1}$  for  $1 \leq j \leq L-1$ , and that located at the boundary are calculated as follows,

$$U B_{j,j+1}^\dagger U^{-1} = e^{-i\pi/L} a_j^\dagger b_{j+1}^\dagger - e^{i\pi/L} b_j^\dagger a_{j+1}^\dagger, \quad (8)$$

$$U B_{L,1}^\dagger U^{-1} = -(e^{-i\pi/L} a_L^\dagger b_1^\dagger - e^{i\pi/L} b_L^\dagger a_1^\dagger). \quad (9)$$

In the latter case, a negative sign appears for each valence bond. This reflects a property of an  $S = 1/2$  spin which needs  $4\pi$  rotation to return to the original state. Thus the asymptotic form of  $z_L$  is given by

$$z_L^{(1)} = \langle \Psi_{\text{VBS}}^{(m,n)} | U | \Psi_{\text{VBS}}^{(m,n)} \rangle = (-1)^n [1 - \mathcal{O}(1/L)]. \quad (10)$$

It turns out that  $z_L$  changes its sign according to the number of valence bonds at the boundary. The value of  $z_L$  for  $m = n = 1$  was calculated by Totsuka and Suzuki [12]. A similar relation was also found by Bonesteel who discussed two-dimensional  $S = 1/2$  dimer systems [13]. In Eq. (10), the factor  $(-1)^n$  originates from the relative twist angle  $2\pi$  at the boundary, so that it

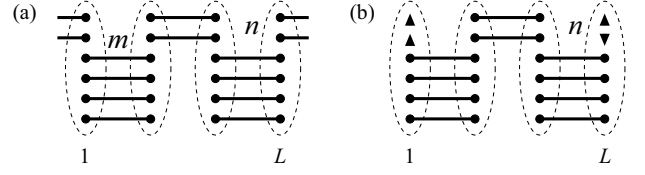


FIG. 1: Generalized VBS states with  $(m, n) = (4, 2)$  for (a) a periodic chain and for (b) an open chain with even  $L$ . Broken ovals mean the symmetrization of  $S = 1/2$  variables at each site, and solid triangles denote isolated bosons.

does not depend on the detailed structure of the twist operator (2). Therefore, the definition of  $z_L$  is still meaningful even in cases with broken translational symmetry ( $m \neq n$ ) which does not accord with the LSM argument. Especially, in the present definition,  $z_L$  has a useful symmetry  $z_L \rightarrow (-1)^{2S} z_L$  under interchange of  $m$  and  $n$ .

In order to calculate  $z_L$  in more detail, we introduce the matrix product (MP) formalism [14, 15]. This is useful to calculate the ground-state expectation values and correlation functions. In this formalism, Eq. (7) is expressed as

$$|\Psi_{\text{VBS}}^{(m,n)}\rangle = \frac{1}{\sqrt{\mathcal{N}}} \text{Tr } g_1^A \otimes g_2^B \otimes \dots \otimes g_{L-1}^A \otimes g_L^B. \quad (11)$$

These matrices for general integer  $S$  case with  $m = n$  are given in Ref. [12]. Based on this result, the matrices for the twisted VBS state  $U^q |\Psi_{\text{VBS}}^{(m,n)}\rangle$  are given by

$$g_{q,j}^A(s, r) = (-1)^{n-s+1} e^{i(n-2s+2)q\pi/L} \times \sqrt{m C_{r-1} n C_{s-1}} (a_j^\dagger)^{m-r+s} (b_j^\dagger)^{n+r-s} |\text{vac}\rangle_j, \quad (12)$$

$$g_{q,j}^B(r, s) = (-1)^{m-r+1} e^{i(m-2r+2)q\pi/L} \times \sqrt{m C_{r-1} n C_{s-1}} (a_j^\dagger)^{m-r+s} (b_j^\dagger)^{n+r-s} |\text{vac}\rangle_j, \quad (13)$$

where  $1 \leq r \leq m+1$ ,  $1 \leq s \leq n+1$ , and  $|\text{vac}\rangle_j$  is the boson vacuum at the  $j$ -th site. By introducing a transfer matrix defined as

$$G_q^A(r_1, r_2; s_1, s_2) \equiv g_{0,j}^{A\dagger}(r_1, s_1) g_{q,j}^A(r_2, s_2), \quad (14)$$

we obtain  $z_L$  as

$$z_L^{(1)} = (-1)^n \frac{\text{Tr } [G_1^A G_1^B]^{L/2}}{\text{Tr } [G_0^A G_0^B]^{L/2}}. \quad (15)$$

Although the analytic form of  $z_L$  is complicated in general, one can evaluate numerical values of Eq. (15), which will be shown in Fig. 2 below. In the fully dimerized case  $(m, n) = (2S, 0)$ ,  $z_L$  is given by a simple form  $z_L^{(1)} = [\sum_{k=0}^{2S} e^{i(2S-2k)\pi/L} / (2S+1)]^{L/2}$ .

One can show that  $z_L$  behaves similarly even in open systems. For even  $L$  (see Fig. 1(b)), the VBS state of Eq. (7) has  $(n+1)^2$ -fold degeneracy due to the isolated bosons at both ends [6]. Then,  $z_L$  changes its sign according to the number of isolated bosons at the  $L$ -th site,

$$z_L^{(1)} = (-1)^n e^{-i\pi/L} [1 - \mathcal{O}(1/L)]. \quad (16)$$

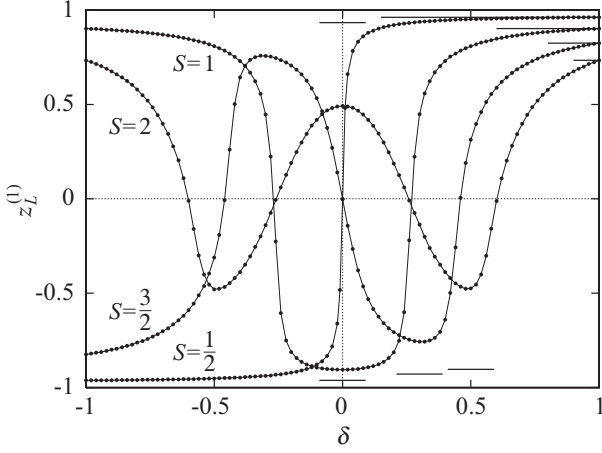


FIG. 2:  $\delta$  dependence of  $z_L^{(1)}$  of the  $L = 64$  periodic BAHC with  $S = \frac{1}{2}, 1, \frac{3}{2}$ , and  $2$ , obtained by the QMC calculation. The horizontal lines indicate  $z_L$  for the VBS states calculated by the MP method [Eq. (15)].

Although  $z_L$  is complex in this case, the imaginary part vanishes in the large- $L$  limit, and the real part of  $z_L$  behaves in the same way as that of the periodic cases. For open systems with odd  $L$ , the number of isolated bosons at the  $L$ -th site is  $m$ , and the ground state has  $(m+1)(n+1)$ -fold degeneracy. Therefore,  $z_L$  is simply given by Eq. (16) multiplied by  $(-1)^{2S}$ .

Next we consider  $z_L$  in connection with a low energy effective theory. According to Schulz's bosonization analysis [16], the Lagrangian density of the uniform Heisenberg spin chain is given by the sine-Gordon model,

$$\mathcal{L} = \frac{1}{2\pi K} [\nabla\phi(x, \tau)]^2 - \frac{y_\phi}{2\pi\alpha^2} \cos[q\sqrt{2}\phi(x, \tau)], \quad (17)$$

where  $\tau$  is the imaginary time,  $\alpha$  is a short range cut off, and  $K$  and  $y_\phi$  are the parameters determined phenomenologically. The phase field is related to that used in Ref. [16] by  $\phi = 2\sqrt{S}\psi_1$ , and  $q = 1$  ( $q = 2$ ) for  $S$  integer ( $S$  half-odd integer), where  $q$  corresponds to degeneracy of gapped ground states. In the gapped (gapless) region one has  $y_\phi(l) \rightarrow \pm\infty$  ( $y_\phi(l) \rightarrow 0$ ) for  $l \rightarrow \infty$  under renormalization  $\alpha \rightarrow e^l\alpha$ . On the unstable Gaussian fixed line [ $y_\phi(0) = 0$  with  $Kq^2 < 4$ ], a second-order ‘‘Gaussian transition’’ takes place between the two gapped states. In this formalism, and the spin wave excitation created by  $U$  corresponds to the vertex operator  $\exp(i\sqrt{2}\phi)$ , so that  $z_L$  for  $q(S-m) = \text{integer}$  is related to the ground-state expectation value of the nonlinear term as  $z_L^{(q)} \propto \langle \cos(q\sqrt{2}\phi) \rangle$  and the three fixed points  $y_\phi = \pm\infty, 0$  correspond to  $z_\infty = \mp 1, 0$ , respectively. Thus the Gaussian critical point can be identified by observing  $z_L^{(q)} = 0$  [17].

In order to demonstrate the above argument, we consider the bond-alternating Heisenberg chain (BAHC),

$$\mathcal{H} = J \sum_{j=1}^L [1 - \delta(-1)^j] \mathbf{S}_j \cdot \mathbf{S}_{j+1}. \quad (18)$$

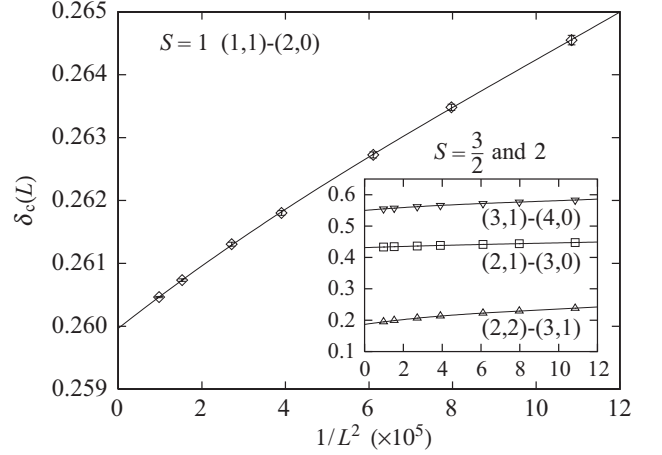


FIG. 3: System size dependence of the successive dimerization transition points  $\delta_c(L)$  of the periodic BAHC with  $S = \frac{1}{2}, 1, \frac{3}{2}$ , and  $2$ . The extrapolation to the  $L \rightarrow \infty$  limit is done by  $\delta_c(L) = \delta_c(\infty) + A/L^2 + B/L^4 + C/L^6$  (solid lines).

For this model, the VBS picture is considered to be realized approximately: The configuration of the valence bonds  $(m, n)$  changes from  $(0, 2S)$  to  $(2S, 0)$  successively as  $\delta$  is increased from  $-1$  to  $1$ , meaning the existence of  $2S$  quantum phase transitions [6, 18]. Since the translational symmetry is explicitly broken, the effective model of the BAHC is given by Eq. (17) with  $q = 1$  for any  $S$  [16], and these transitions are described as a Gaussian type.

In Fig. 2, we show  $z_L$  (with  $q = 1$ ) of the  $L = 64$  periodic BAHC with  $S = \frac{1}{2}, 1, \frac{3}{2}$ , and  $2$  as a function of  $\delta$ . For the calculation of  $z_L$ , we employ the QMC method with the continuous-time loop algorithm [19]. We used the multi-cluster variant of the loop algorithm. The QMC steps are  $10^3$  for thermalization, and  $5 \times 10^5 - 10^6$  for the measurement, and the inverse temperature  $\beta$  is taken large enough so that the value of  $z_L$  can be identified as that of the ground state. The largest inverse temperature used in this calculation is  $\beta J = 128$ .

As seen clearly in Fig. 2, the  $\delta$  dependence of  $z_L$  agrees qualitatively with the present interpretations based on the VBS picture, which predicts  $z_\infty = (-1)^n$  with a symmetry  $z_L \rightarrow (-1)^{2S} z_L$  for  $\delta \leftrightarrow -\delta$  ( $m \leftrightarrow n$ ). We have also calculated  $z_L$  for open chains with even and odd  $L$ 's, and confirmed that the results agree with our predictions including Eq. (16). In Fig. 2, we also present  $z_L$  for the VBS states calculated by the MP method [Eq. (15)] as horizontal lines. The difference between  $z_L$  for the BAHC and that for the VBS states becomes larger as  $S$  increases. This means that the VBS picture becomes poor for the BAHC with large  $S$ .

Next, we determine the critical point  $\delta_c$  by observing  $z_L = 0$  with  $L$  up to 320 (Fig. 3). In this calculation, the inverse temperature is taken as  $\beta J = L/2S$ , being assumed the Lorentz invariance. Extrapolation to

the  $L \rightarrow \infty$  limit has been done by the least-squares fitting by assuming the asymptotic form as  $\delta_c(L) = \delta_c(\infty) + A/L^2 + B/L^4 + C/L^6$ . This polynomial with even powers of  $1/L$  is justified by the parity symmetry which ensures that  $z_L$  is real [Eq. (5)]. For  $S = 1$ , we have obtained the critical value for the transition between the (1,1) and (2,0) phases as  $\delta_c = 0.25997(3)$ , where  $()$  denotes  $2\sigma$ . This result is consistent with the previous estimates:  $\delta_c = 0.2595(5)$  by the QMC calculation for the susceptibility [20] and 0.2598 by the level-crossing method [21]. Similarly, we identify the critical point of the  $S = 3/2$  case [(2,1)-(3,0)] and those of the  $S = 2$  case [(2,2)-(3,1), (3,1)-(4,0)]. We obtain  $\delta_c = 0.43131(7)$ , 0.1866(7), and 0.5500(1), respectively (see the inset of Fig. 3). They are also consistent with  $\delta_c = 0.4315$ , 0.1830, and 0.5505 obtained by the level-crossing method [22], but much more accurate.

Here, we comment on the method used in Refs. [21] and [22] proposed by Kitazawa who pointed out that the Gaussian transition with  $q = 1$  can be identified by a level crossing of excitation spectra under twisted boundary conditions [23]. Since this method is also explained by the sine-Gordon theory [23] and the VBS picture [21, 22], the results are considered to be equivalent to ours. Although finite-size corrections in the level-crossing point in VBS states tend to be smaller than those of the  $z_L = 0$  point, the application of the level-crossing method to larger systems is difficult, because it relies on the exact diagonalization to obtain excitation spectra which needs the whole Hilbert space. On the other hand, our approach is based only on the ground state quantity,  $z_L$ , so that various numerical methods such as the present QMC method can be employed. This makes it possible to deal with systems with enormous Hilbert space, such as large- $S$  and ladder systems. Note that the density matrix renormalization group method is also suitable for our approach, since  $z_L$  remains as a meaningful order parameter even in open systems. Furthermore, our method also has an advantage to comprehend the physical picture as already discussed.

Finally, we discuss the relation between  $z_L$  and the string order parameter. In Ref. [6], Eq. (1) for  $|\Psi_{\text{VBS}}^{(m,n)}\rangle$  was calculated as  $\mathcal{O}_{\text{string}}^z = [\frac{m+n+2}{2(m+2)}]^2 \delta_{n,\text{odd}}$ , where finite and vanishing  $\mathcal{O}_{\text{string}}^z$  correspond to negative and positive  $z_L$ , respectively. However, this calculation is limited only for  $S$  integer, and the twist angle  $\pi$  in Eq. (1) is needed to be generalized for other cases [6]. In addition, it is difficult to determine accurate phase transition points by  $\mathcal{O}_{\text{string}}^z$ , because it does not change the sign. Thus  $z_L$  turns out to be more rational order parameter to describe VBS ground states in a unified way.

In summary, we have introduced  $z_L$  given as the ground-state expectation value of the twist operator (3) as an order parameter to characterize various ground states in quantum spin chains. Especially,  $z_L$  changes

the sign according to the configuration of valence bonds. This property enables us to determine the critical point between different VBS states by observing  $z_L = 0$ . We have demonstrated this theory by using the QMC simulation for the successive dimerization transitions of the BAHc, and determined the phase boundaries with quite high accuracy.

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